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Dermal & inhalation exposure of operators during fungicide application in vineyards. Evaluation of coverall performance

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HIGHLIGHTS

- Operator exposure was measured during fungicide hand-held application in vineyards.
- The potential exposure of body and hands accounted for 89% and 9%, respectively.
- Total actual dermal exposure ranged from 2 to 19 mg/kg active substance applied.
- The German model overestimates the body actual dermal exposure (75th percentiles).
- The two protective coveralls tested provided satisfactory protection to operators.

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ABSTRACT

In the present study the dermal and the inhalation exposure of five operators during fungicide applications in vineyards were determined. The produced exposure datasets can be used as surrogate for the estimation of the actual and the potential dermal as well as inhalation operator exposure levels for this application scenario. The dermal exposure was measured using the whole body dosimetry method while the inhalation exposure with the use of personal air sampling devices with XAD tubes located on the operator's breathing zone. Ten field trials were carried out by 5 different operators using a tractor assisted hand-held lance with spray gun at the Tanagra region of Viotia, Greece. An in-house GC–ECD analytical method was developed and validated for the determination of penconazole, which was the active substance (a.s.) of the fungicide formulation used in field trials. The mean recovery of field-fortified samples was 81%. The operator exposure results showed expected variability and were compared to those derived from the German model for prediction of operator exposure. The comparison of the 75th percentile values for an operator wearing personal protection equipment has shown that the measured levels were 2.2 times lower than those estimated by the German model. The levels of actual dermal exposure ranged from 2 to 19 mg/kg a.s. applied. The protection provided by the two types of coveralls was evaluated and in comparison to the existing reduction factors used for other types of PPE (coveralls) was found satisfactory for the operator under the conditions of the specific applications.
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1. Introduction

Based on the EU Legislation for the placing of plant protection products (PPPs) on the market, a risk assessment must be carried out for all possible exposure scenarios of operators, workers, residents and bystanders that can be expected to occur as a consequence of the proposed uses of a PPP.

The Regulation (EC) 1107/2009 has introduced the zonal approach in the evaluation and registration of PPPs. EU is divided in three geographical zones, meaning in practice that regulators and risk managers need to take into account regional differences (like environmental conditions, application techniques, etc.) when performing the assessment of exposure to PPPs. Greece belongs to the Southern EU zone.

Grapevine is one of the most demanding crops with respect to its plant protection needs. Operators apply PPPs in vineyards, up to 15 times per year for the control of various pests and diseases (Hocking et al., 2007). This fact underlines the necessity for efficient protection of the operators as well as for reliable risk assessment.

One of the most effective approaches to control the undesirable effects of any substance is to minimize the exposure of humans and other non-target organisms (Hatcher et al., 2008; McKinlay et al., 2008; Nasterlack, 2007; Porta et al., 2008a, 2008b). The proper use of personal protective equipment (PPE) can drastically reduce the operator exposure levels to

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PPPs (van der Jagt et al., 2004). Operators applying PPPs are considered to be the group exposed to the highest levels of PPPs. Although this exposure is a consequence of occupational activities, the protection measures taken are not always reflecting professional approaches (Glass and Machera, 2009). In addition skin is the main route of exposure during application, thus the evaluation of PPE performance for reducing especially skin exposure to pesticides is essential (Protano et al., 2009).

The determination of the exposure levels is a substantial step in the decision-making procedure for reliable risk assessment for operators, workers, bystanders, consumers and the environment. Operator exposure levels can be either estimated using the available calculation models such as the German model and the UK POEM or measured under real application conditions. Both methods have their advantages and disadvantages. The calculation models have been based on old and limited field trials that that have been conducted in northern European conditions. It is noted that the estimated levels of operator exposure differ significantly depending on the predictive model applied. Biomonitoring studies can also provide useful information regarding pesticide residues and their metabolites in biological fluids. However, biomonitoring results are not regularly used for risk assessment purposes due to lack of detailed data that would allow the reliable extrapolation to dermal exposure levels.

The objective of the present study is to measure the operator exposure levels during fungicide application in vineyards under the currently followed application practices in Greece. These datasets can operate as surrogates for the estimation of the actual and the potential dermal exposure and the inhalation exposure levels during fungicide applications for the specific application scenario. Furthermore, the field performance of different types of protective coveralls is evaluated.

2. Materials and methods

2.1. Field phase

The study was conducted in the grapevine growing area of Tanagra, Viotia prefecture, Greece which is very well known for wine production since the ancient times, being also the country of ancient Greek poet Hesiod who is considered by many historians as the «father of viticulture».

The vineyards sprayed in the frame of the study covered a total area of 3.6 ha (approx. 0.3–0.4 ha/vineyard). The vineyards had an average crop height of 1.2 m. The average planting distance between rows was 2.4 m. The cordons (arms) of the grapevines were trained horizontally along wires resembling the letter “T”, corresponding to a typical vineyard crop of Greece. Detailed data on the crop parameters and field conditions during the application are provided for each vineyard in Table 1, whereas in Photo 1 an operator of the study applying fungicide to a vineyard is depicted corresponding to the respective typical application scenario. This scenario may be considered representative for southern European countries in cases where hand-held spraying is applied.

The procedure followed was based on the principles of the whole body dosimetry method (Chester, 1993; Machera et al., 2003; OECD, 1997; WHO, 1982) as it has been adapted and described in detail in our previous study (Tsakirakis et al., 2011). Two coverall types were used, type A coverall (50/50 cotton/polyester with Resist Spills® finish) and type B coverall (100% cotton), serving as both operator's PPE and external exposure dosimeters. Inner dosimeters (100% cotton shirts and long pants) were also used for the monitoring of actual exposure. Head and hand contamination as well as inhalation exposure were measured in accordance to the sampling method already used in a previous study (Tsakirakis et al., 2011) using as dosimeter caps, gloves and personal air samplers respectively.

A total of 5 operators with adequate experience in PPP applications participated in the study. Each of them carried out two applications (one per coverall type) while none of them was involved in mixing and loading procedure. A 10% EC formulation of penconazole was used as representative fungicide and was applied according to the label recommended dose rate (0.040 g a.s./L). The application technique, hand-held single nozzle spray guns connected to a tractor tank, was the one usually followed in the specific vineyard. All operators were instructed to follow their normal spraying practice. The duration of applications was approximately 2 h, corresponding to a treated area of 0.3–0.4 ha, which is also typical for the vineyard farms in the region. At the end of the application period, the dosimeters were transferred to freezer within 2 h from the termination of each application day. Special care was taken to avoid any cross contamination of the dosimeter samples. Details of the operators, application conditions and parameters are shown in Table 1. Quality control samples of all dosimeters used were fortified in the field as a measure of the active substance (a.s.) stability and recovery according to previously described procedure (Machera et al., 2009; Tsakirakis et al., 2011). Satisfactory results were obtained at all fortification levels with recoveries, for low and high concentrations, well above the cut-off value of 70% (OECD, 1997). Relative standard deviation (%RSD) values for both low and high concentrations were <4% with acceptable limit being <20% (OECD, 1997). The corresponding field fortification volumes, recovery and RSD values are presented in Table 2.

2.2. Dosimeter residues & expression of exposure

The total amount of active substance penconazole detected on the outer and on the inner dosimeters corresponds to potential exposure, while the respective amount of a.s. detected on the inner dosimeter represents actual exposure. The total potential dermal exposure (total PDE) of the operator is the sum of the potential exposure for the body (i.e. trunk inner + trunk outer dosimeters) plus the potential exposure for the hands (i.e. inner gloves + outer gloves) plus the head exposure. Respectively the total actual dermal exposure (total ADE) of the operator is the sum of the actual exposure for the body (i.e. trunk inner dosimeters) plus the actual exposure for the hands (i.e. inner gloves) plus the head exposure.

The hand exposure was measured, as mentioned above, from the a.s. residues found on the gloves with the inner glove residues corresponding to the actual hand exposure and the sum of a.s. measured on the outer (protective) and inner gloves corresponding to the potential hand exposure.

For the operator's head, since no means of PPE was used, the potential and actual head exposure coincide corresponding to the amount of active substance measured on the operator's cap multiplied by a factor of 2.

2.3. Analytical method

The active substance (a.s.) used for the method validation was penconazole and the respective analytical standard was 99.1% pure and was purchased from Riedel de Haën (Buchs, Switzerland). Stock solution of the reference item was prepared at 500 μg/mL and the respective working solutions of 0.5, 1, 2, 5 and 20 μg/mL by further dilution. Triphenylphosphate (purity 98%) was obtained from Fluka (Buchs, Switzerland) and used as internal standard (0.25 μg/mL). The solvent used for the preparation of all the above solutions as well as for the extraction of penconazole from different matrices was *n*-hexane (Merck, Darmstadt Germany). All solutions were stored at −18 °C.

The analytical method was developed and validated in the laboratory (Goumenou and Machera, 2001; Machera et al., 2001). The residues of the a.i. penconazole were extracted from the samples with n-hexane following the procedure described in our previous study (Tsakirakis et al., 2011). Especially for the outer gloves (nitrile) the extraction had been already carried out in the field after the end of each application following the procedure described in the aforementioned study since it was known that recovery of the a.s. reduces over time with this matrix (Durham and Wolfe, 1962).

Penconazole concentration in extracts was adjusted to be in the range of linear responses of the gas chromatographic detector by further

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